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Optimization of torrefaction conditions for high energy density solid biofuel from oil palm biomass and fast growing species available in Malaysia

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ABSTRACT

Without appropriate treatment, lignocellulosic biomass is not suitable to be fed into existing combustion systems because of its high moisture content, low bulk energy density and difficulties in transport, handling and storage. The aim of this study was to investigate the effects of torrefaction treatment on the weight loss and energy properties of fast growing species in Malaysia (*Acacia* spp., and *Macaranga* spp.) as well as oil palm biomass (oil palm trunk and empty fruit bunch). The lignocellulosic biomass was torrefied at three different temperatures 200, 250 and 300 °C for 15, 30 and 45 min. Response surface methodology was used for optimization of torrefaction conditions, so that biofuel of high energy density, maximized energy properties and minimum weight loss could be manufactured. The analyses showed that increase in heating values was affected by treatment severity (cumulated effect of temperature and time). Our results clearly demonstrated an increased degradation of the material due to the combined effects of temperature and treatment time. While the reaction time had less impact on the energy density of torrefied biomass, the effect of reaction temperature was considerably stronger under the torrefaction conditions used in this study. It was demonstrated that each biomass type had its own unique set of operating conditions to achieve the same product quality. The optimized torrefaction conditions were verified empirically and applicability of the model was confirmed. The torrefied biomass occurred more suitable than raw biomass in terms of calorific value, physical and chemical properties. The results of this study could be used as a guide for the production of high energy density solid biofuel from lignocellulosic biomass available in Malaysia.

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1. Introduction

Biomass is a primary source of renewable carbon that can be utilized as a feedstock for biofuels production in order to achieve energy independence. As one of the largest producers and exporters of palm oil, Malaysia has abundant availability of oil palm biomass as a promising source of lignocellulosic raw material. Additional quantities of lignocellulosic biomass can be yielded from energy crops. These crops are fast-growing plants or trees which are harvested specifically for energy production (Sims et al., 2006). In nature, lignocellulosic biomass has high moisture content, low bulk energy density and is difficult to transport, handle, store and feed into existing combustion systems without pretreatments. When lignocellulosic biomass is employed for bioenergy, pretreatments of lignocellulosic biomass are essential procedures for achieving higher efficiencies of fuel production or consumption (Tabil et al., 2011). For example, moisture content from the lignocellulosic biomass was driven off to enhance the combustion efficiency and prevent microbial degradation during storage (Chin et al., 2012). While, in liquid biofuel production, acid pretreatments have been adopted widely to facilitate the conversion of hemicelluloses and cellulose into soluble sugars for lignocellulosic ethanol production (Lloyd and Wyman, 2005; Chin et al., 2010, 2011).

In addition to the aforementioned methods, torrefaction of biomass is another notable pretreatment method that enables energy densification of biomass and biomass homogenization (Yan et al., 2009; Arias et al., 2008). This pretreatment has been recommended as an efficient way to enhance solid biofuel properties by water removal, reduction of the hygroscopic range, and increased grindability. Torrefaction is a thermal pretreatment process by

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Table 1
Proportion of bark on the stem and basic density of wood.

Lignocellulosic biomass	Proportion of bark (wt%)	Density (kg/m^3)
Acacia spp.	9.48	494
Macaranga spp.	12.13	371
Oil Palm Trunk (OPT)	–	398

subjecting wood to temperature levels between 200 and 300 °C in the absence of oxygen. During this process, the cell walls are degraded and the nature of the resulting product lies between that of wood and charcoal. The residual product is in solid form, which is referred to as torrefied biomass. Torrefaction is influenced by many parameters including the composition and physical properties of the biomass and operating condition (Arias et al., 2008; Esteves and Pereira, 2009).

Thus, the purpose of this study was to build a model of torrefaction procedure applicable in production of solid biofuel from lignocellulosic biomass available in Malaysia (fast growing species and oil palm biomass). Changes in the energy properties of the lignocellulosic biomass when subjected to torrefaction in the temperature range of 200–300 °C with different treatment time were investigated. Weight loss is an important parameter for optimizing the design and operation of a biomass torrefaction plant. So that understanding of the process variables for optimization is required. The present study using the technique of response surface methodology (RSM) enables finding the optimum torrefaction conditions for a high density solid fuel. Another important research parameter is the type and chemical composition of the biomass used, which determine its behavior in the torrefaction process. Therefore, a set of chemical analyses was performed to quantify the alteration in extractives, acid insoluble materials, holocellulose and cellulose in the lignocellulosic biomass, in order to better understand these modifications and eventually to define a predictive approach for other species. This study is therefore devoted to the modification of energy characteristic of selected fast growing species grown in Malaysia (*Acacia* spp., and *Macaranga* spp.) and oil palm (*Elaeis guinesis*) biomass (oil palm trunk and empty fruit bunch) subjected to different torrefaction treatments.

2. Materials and methods

2.1. Material selection preparation

The oil palm biomass used in this study includes oil palm trunk (OPT) and empty fruit bunch (EFB). The selected fast growing species were *Acacia* spp., and *Macaranga* spp. with the trunk diameter range between 10 and 15 cm. For the fast growing species, the samples were mixtures of xylem and bark. Densities of OPT and fast growing species were determined by the water displacement volume method. The wood-bark ratios were obtained by removing the bark from the logs and the samples (wood and bark) were then oven dried and weighed. The ratio in percentage was calculated. The bark content and density of the fast growing species and oil palm trunk are shown in Table 1. All types of woody material (fast growing species, oil palm trunk and empty fruit bunch fines) were ground and sieved to obtain 40-mesh fine particles.

2.2. Torrefaction process

The ground wood material was dried at 105 °C for 24 h before torrefaction in order to remove the residual water remaining in the biomass. The dried wood fine (50 g) was placed in a furnace and torrefied under the conditions as follows: temperature 200 °C, 250 °C and 300 °C for 15 min, 30 min and 45 min. Torrefaction was performed in a tube furnace controlled by a continuous nitrogen flow rate (0.5–1.0 L/min) to remain the oxygen level below 1%. The oxygen level was measured on the gas flow leaving the furnace using a portable gas sensor. For each treatment, the weight loss of the torrefied wood was calculated using the following expression (Eq. (1)).

$$W_L = \left[\frac{M_I - M_D}{M_I}\right] \times 100 \quad (1)$$

where W_L is the weight loss (%), M_I is the initial oven-dried mass of the lignocellulosic material and M_D is the oven dried mass of the wood sample after torrefaction.

2.3. Determination of chemical composition

Wood composition was performed according to TAPPI standard T 203. The extractives content, acid-insoluble materials (lignin), holocellulose content and alphacellulose content were determined on untreated and torrefied samples. The extractives content was determined as the content of products soluble in acetone and ethanol with a ratio of 2:1. The acetone/ethanol extraction was performed under reflux for 5 h. The extractives obtained were evaporated at 40 °C using a rotary evaporator to remove most of solvent, and dried in an oven at 35 °C for 24 h. Acid-insoluble materials consisting by a majority of lignin, were determined by quantitative acid hydrolysis with 72% H_2SO_4 according to the T-249 em-85. Holocellulose and alphacellulose was determined according to T 429 cm-10, Wise method (Wise et al., 1946). The ash content was determined by burning (dry oxidation) the oven-dried sample (2 g) in a muffle furnace model at 575 ± 25 °C for 4 h as per the TAPPI standard method, T211 om-85. This test method was used to determine the amount of ash, remaining after dry oxidation of the sample. All analyses were performed in triplicate and the mean were presented.

2.4. Determination of higher heating value (HHV)

The calorific value was determined according to BSI standard EN 14918 with a bomb calorimeter, in which 0.50 g of oven-dried biomass was completely combusted under a pressurized (3000 kPa) oxygen atmosphere.

2.5. Surface response analysis

Second-degree polynomials were calculated using Design-Expert Version 8.0.1 software (Stat-Ease Inc., USA) to estimate the response of the dependent variables. Temperature (X_1, 200–300 °C) and reaction time (X_2, 15–45 min) were chosen as the independent variables. HHV yield of lignocellulosic biomass was used as the dependent output variables.

3. Results and discussion

3.1. Chemical composition

The change in the chemical composition (extractives, acid insoluble fraction, holocellulose and cellulose) of torrefied lignocellulosic biomass is shown in Table 2. The two fast growing species and EFB showed a similar variation in extractives content after the treatments. During mild torrefaction (below 250 °C), the extractives content tented to increase, whereas with a more severe condition (above 250 °C), the extractives content decreased. According to Weiland and Guyonnet (2003) the increase in the extractives content is observed probably due to the onset of hemicelluloses degradation, which produces new extractible components. At high temperature levels, the decrease in extractive content might result from its thermal degradation. However, OPT

Table 2
Chemical composition of torrefied lignocellulosic biomass.

Temp (°C)	Reaction time (min)	Extractives (%)	Acid-insoluble (%)	Holocellulose (%)	Cellulose (%)	Ash (%)
Acacia spp.						
	Untreated	2.72	23.69	71.69	43.16	1.71
200	15	3.71	26.36	68.12	44.45	1.75
200	30	4.05	27.70	66.19	45.02	2.04
200	45	4.35	28.12	65.39	46.09	2.05
250	15	4.01	28.44	64.90	49.33	1.77
250	30	3.76	29.50	64.82	52.20	1.90
250	45	3.36	30.41	63.37	51.65	2.25
300	15	3.20	33.50	61.32	49.77	1.90
300	30	2.38	42.04	53.32	46.83	2.22
300	45	1.97	44.20	51.20	45.41	2.43
Macaranga spp.						
	Untreated	2.06	27.03	68.74	45.64	2.10
200	15	2.19	28.07	67.06	48.00	2.62
200	30	2.65	29.34	64.74	49.02	3.19
200	45	2.8	29.74	65.54	48.91	3.34
250	15	3.10	31.05	62.9	50.45	2.78
250	30	3.46	32.61	60.42	49.58	3.33
250	45	3.86	35.14	57.46	48.48	3.37
300	15	3.01	38.42	55.23	46.52	2.91
300	30	2.84	41.91	51.80	43.86	3.38
300	45	2.02	44.04	50.11	42.81	3.47
EFB						
	Untreated	6.37	12.69	74.85	44.24	5.96
200	15	7.13	14.14	72.42	45.00	6.23
200	30	7.13	14.52	72.06	45.63	6.25
200	45	7.71	15.89	70.02	47.17	6.26
250	15	7.60	16.21	69.79	48.71	6.35
250	30	7.13	18.20	67.35	46.39	6.75
250	45	6.37	22.05	64.24	45.57	7.06
300	15	6.82	23.28	63.22	47.10	6.61
300	30	6.72	24.57	61.81	46.35	6.83
300	45	5.47	31.95	54.43	37.15	7.30
OPT						
	Untreated	6.78	10.84	73.65	33.44	1.33
200	15	6.84	11.12	73.50	38.06	1.32
200	30	6.60	11.89	75.97	46.98	1.41
200	45	5.54	12.82	74.69	45.34	1.88
250	15	5.38	12.07	74.15	46.14	1.36
250	30	5.16	14.41	71.53	44.11	1.54
250	45	4.84	16.82	70.13	42.76	1.88
300	15	4.22	19.55	69.19	42.01	1.41
300	30	4.03	23.87	66.23	39.96	1.89
300	45	3.76	27.95	59.95	36.76	2.05

showed a different trend as the extractives content decreased even at the mildest torrefaction temperature of 200 °C. It has been reported by H'ng et al. (2013) that OPT contained as much as 6% of starch which is soluble in alcohol–acetone mixture. Mano et al. (2003) also stated that the degradation process of starch initiated at 150 °C. When torrefaction treatment is applied on OPT, starch might be degraded causing the overall reduction of extractives content.

Our data indicate a dramatic reduction of holocellulose content with increasing severity of torrefaction (Table 2). Hemicelluloses, cellulose and lignin are known to decompose at temperature of 225–300 °C, 305–375 °C and 250–500 °C, respectively (Prins et al., 2006). It is apparent that holocellulose is significantly affected by the torrefaction, even at the temperature as low as 200 °C in all cases. Holocellulose consists of hemicellulose and cellulose. Cellulose content in the biomass is increasing when mild torrefaction treatment is applied. This might be due to severe degradation of hemicelluloses that contributed to the increase in cellulose content in the biomass. According to the literature, the decrease in holocellulose is due to the reduction of hemicelluloses, which begins to be significant at temperature above 180 °C (Rousset et al., 2011; Chen and Kuo, 2011). On the contrary, cellulose is thermally more stable. Our results showed that cellulose was influenced markedly only when severe torrefaction (above 250 °C) was applied. Chen and Kuo (2011) stated that cellulose degradation starts at about 230 °C but becomes significant only at temperature levels above 290 °C.

Acid-insoluble fraction consisting by a majority of lignin was not degraded under the torrefaction conditions used in this study (Table 2). Acid-insoluble material content increased linearly in all cases with an increase in weight loss due the thermal treatment. This was mainly due to a side-effect of the decomposition of hemicelluloses and to a lesser extent, due to cellulose content. As a result, lignin was accumulated in the torrefied biomass, and acid-insoluble material content increased. Alternatively, according to Sannigrahi et al. (2011), some products derived from the degradation of carbohydrates could be formed and increase the acid-insoluble fraction content. Hemicelluloses are more sensitive to thermal degradation giving more degradation products such as furfural (Kamden et al., 2002). This molecule can react with lignin phenolic compounds, giving some acid-insoluble compounds. Agrawal (1988) found that low temperatures of pyrolysis (below 280 °C) yielded scission reactions, which favor crosslinking, as well as aromatization reactions leading to formation of a solid product (charcoal) having high content of acid-insoluble fractions. Among the fast growing species, *Acacia* spp. has the highest increment of acid-insoluble material (21% increment) subjected to severe torrefaction (300 °C for 45 min). This was associated with the highest holocellulose degradation (20% decrease) found in *Acacia* spp. which contributed to the increment of acid-insoluble fraction. This result is very similar to that reported by Phanphanich and Mani (2013).

Table 3
Weight loss and calorific value of torrefied lignocellulosic biomass.

Temperature (°C)	Reaction time (min)	Weight loss (%)	HHV (MJ/kg)	HHV yield (%)
Acacia spp.				
Untreated		0	18.35	100
200	15	3.23	18.62	98.19
200	30	3.67	19.16	100.58
200	45	5.37	20.42	105.30
250	15	4.48	24.15	125.71
250	30	6.93	25.83	131.00
250	45	11.89	27.53	132.19
300	15	13.26	27.58	130.37
300	30	19.58	27.80	121.84
300	45	28.26	28.94	113.14
Macaranga spp.				
Untreated		0	17.95	100
200	15	0.90	18.08	99.82
200	30	1.55	19.45	106.65
200	45	3.45	20.05	107.84
250	15	2.87	21.50	116.30
250	30	4.28	23.78	126.78
250	45	6.60	25.27	131.46
300	15	10.72	26.03	129.43
300	30	15.26	27.16	128.21
300	45	20.51	28.79	127.51
EFB				
Untreated		0	18.07	100
200	15	4.71	21.01	110.79
200	30	7.24	23.02	118.17
200	45	9.65	23.80	119.04
250	15	8.79	22.24	112.30
250	30	9.89	24.17	120.54
250	45	10.95	24.54	120.98
300	15	11.13	22.63	111.34
300	30	19.08	24.34	109.03
300	45	25.31	25.03	103.46
OPT				
Untreated		0	17.18	100
200	15	6.19	17.54	95.78
200	30	6.74	17.62	95.64
200	45	7.68	17.66	94.88
250	15	7.95	17.79	95.30
250	30	8.05	17.96	96.09
250	45	8.85	18.18	96.43
300	15	8.81	18.27	96.96
300	30	11.55	21.00	108.10
300	45	15.07	21.89	108.21

3.2. Weight loss and HHV of torrefied lignocellulosic biomass

The HHV and weight loss depended on the torrefaction conditions listed in Table 3. The weight loss increased with time and temperature of treatment for the lignocellulosic material used in this study. Since weight loss is a cumulated effect of reaction time and temperature, it seems to be a good estimator of torrefaction extent (Almeida et al., 2010). A significant difference was apparent among the two fast growing species; the weight loss for *Acacia* spp. was always higher for a given treatment level. For example, the more severe treatment (300 °C; 45 min) caused weight loss of 28.26% for *Acacia* spp. compared with 20.51% for *Macaranga* spp. The higher weight loss of *Acacia* spp. may be caused by the higher degradation rate of the constituents which is clearly shown in Table 2.

The treatment time always had a considerable impact on the weight loss, the sole exception being in the case of OPT at 250 °C, for which the additional reaction time only slightly increased the weight loss (7.95% after 15 min and 8.85% after 45 min). However, increasing temperature had a significant effect on the thermal decomposition of all the lignocellulosic biomass used in this study. At torrefaction temperature of 300 °C, drastic reductions of weight losses were observed. It has been found that the impact of torrefaction on oil palm biomass was more apparent than the selected fast growing species at low torrefaction temperature. At 200 °C and 45 min reaction time, there were only slight reductions in average mass of 4.72% for the two species while the reduction for EFB and OPT were 9.65% and 7.68%, respectively. While, at temperatures of 300 °C and 45 min reaction time, the weight loss for *Acacia* spp. was 28.26% while the weight loss for EFB, OPT and *Macaranga* spp. were respectively, 25.31%, 15.07% and 20.51%.

Higher heating value (HHV) represents the maximum amount of energy embedded per gram of a material. It is important to mention that higher heating value increases with the extent of treatment. The energy value ranged from 18.08 to 28.94 MJ/kg depending on the torrefaction conditions. This implies that the HHV in the torrefied biomass increased by 0.37–67.9% when compared to the untreated material. At temperatures of 300 °C and 45 min reaction time, the HHV increment for *Acacia* spp., *Macaranga* spp. and EFB were respectively 68%, 60%, and 39%, while that for OPT was only 27%. The HHV was affected more by the temperature than by the reaction time. The HHV significantly increased with a torrefaction temperature exceeding 250 °C, regardless of the reaction time. HHV slowly increased as the temperature raised until 250 °C, then dramatically increased as the temperature further increased from 250 °C to 300 °C. It is clear that the characteristics of thermal degradation of the three basic constituents of biomass are different. Furthermore, from the viewpoint of practical applications of torrefaction, if the light torrefaction (200 °C) is applied for the pretreatment of biomass, its main function will be to remove moisture and light volatiles from the biomass (Pierre et al., 2011). Therefore, it is inferred that the heating value of the torrefied biomass is improved to a small extent. If biomass is pretreated using the severe torrefaction conditions (above 300 °C), the role of lignin (acid-insoluble fraction) in energy contribution might be more important than that of holocellulose resulting from a large portion of holocellulose being consumed in the severe torrefaction. High lignin content in torrefied biomass is associated with high energy density due to the ether and carbon—carbon linkages in lignin, which has higher energy than the C—O or C—H bonds (Lehtikangas, 2001; Shafizadeh et al., 1976). Therefore, high lignin content in the torrefied biomass results in a higher HHV.

The highest energy value was 28.94 MJ/kg for *Acacia* spp. torrefied at 300 °C for 45 min, corresponding to a weight loss of 28.26%. An overall increase in calorific value (1–39% and 1–27%) and decrease in weight (5–25% and 6–16%) for EFB and OPT, respectively were observed with the increase in process reaction time and temperature. EFB undergoes greater alterations than OPT in terms of calorific value. In addition, EFB underwent greater changes in terms of weight loss. The obtained relationships allow quantification of the increase in wood energy concentration due to the torrefaction.

Table 4
The mathematical models derived from the experimental results for HHV yield.

Lignocellulosic biomass	Model calculated for HHV yield (Y_1)
Acacia spp.	$Y_{1(A)} = -397.92 + 3.99X_1 - 0.17X_2 + 2.21X_1X_2 - 7.68X_1^2$
Macaranga spp.	$Y_{1(M)} = -185.22 + 2.06X_1 + 1.457X_2 - 2.34X_1X_2 - 3.53X_1^2 - 9.99X_2^2$
EFB	$Y_{1(E)} = -64.08 + 1.27X_1 + 2.23X_2 - 5.38X_1X_2 - 2.39X_1^2 - 0.01X_2^2$
OPT	$Y_{1(O)} = 194.31 - 0.83X_1 - 0.39X_2 + 4.79X_1X_2 + 1.53X_1^2 - 0.01X_2^2$

3.3. Response surface analysis for HHV yield

The primary goal of torrefaction is to refine raw biomass to an upgraded solid fuel, of improved handling qualities and enhanced

Table 5
Analysis of variance (ANOVA) for the adjusted model for the HHV yield of lignocellulosic biomass during torrefaction.

Source	Sum of squares	Degrees of freedom	Mean square	*F*-Value	*p*-Value ((Prob. > F)
Acacia spp.					
Model	4670.04	5	934.01	1360.52	<0.0001
Residual	14.42	21	0.69		
Lack of fit	8.89	3	2.96	9.65	<0.0005
Pure error	5.53	18	0.31		
Corrected total	24,684.46	26			
Temp	2212.81	1	2212.81	3223.28	<0.0001
Time	211.91	1	211.91	308.68	<0.0001
Temp × time	33.03	1	33.03	48.11	<0.0001
$(\text{Temp})^2$	2210.30	1	2210.30	3219.63	<0.0001
$(\text{Time})^2$	1.99	1	1.99	2.90	0.1036
					Adjusted *R*-square: 0.9962
Macaranga spp.					
Model	2606.11	5	521.22	62.23	<0.0001
Residual	175.89	21	8.38		
Lack of fit	174.36	3	58.12	686.39	<0.0001
Pure error	1.52	18	0.085		
Corrected total	2782.00	26			
Temp	2012.60	1	2012.60	240.29	<0.0001
Time	191.14	1	191.14	22.82	<0.0001
Temp × time	4.23	1	4.23	0.50	<0.4853
$(\text{Temp})^2$	385.22	1	385.22	45.99	<0.0001
$(\text{Time})^2$	12.92	1	12.92	1.54	0.2279
					Adjusted *R*-square: 0.8973
EFB					
Model	792.46	5	158.49	42.38	<0.0001
Residual	78.53	21	3.74		
Lack of fit	77.96	3	25.99	816.72	<0.0001
Pure error	0.57	18	0.032		
Corrected total	870.99	26			
Temp	292.02	1	292.02	78.09	<0.0001
Time	40.56	1	40.56	10.85	0.0035
Temp × time	195.05	1	195.05	52.16	<0.0001
$(\text{Temp})^2$	213.32	1	213.32	57.04	<0.0001
$(\text{Time})^2$	51.51	1	51.51	13.77	0.0013
					Adjusted *R*-square: 0.8864
OPT					
Model	672.60	5	134.52	25.53	<0.0001
Residual	110.66	21	5.27		
Lack of fit	84.94	3	28.31	19.81	<0.0001
Pure error	25.73	18	1.43		
Corrected total	783.26	26			
Temp	307.04	1	307.04	58.27	<0.0001
Time	85.01	1	85.01	16.13	0.0006
Temp × time	195.05	1	154.77	29.37	<0.0001
$(\text{Temp})^2$	213.32	1	88.47	57.04	0.0005
$(\text{Time})^2$	51.51	1	37.30	13.77	0.0146
					Adjusted *R*-square: 0.8251

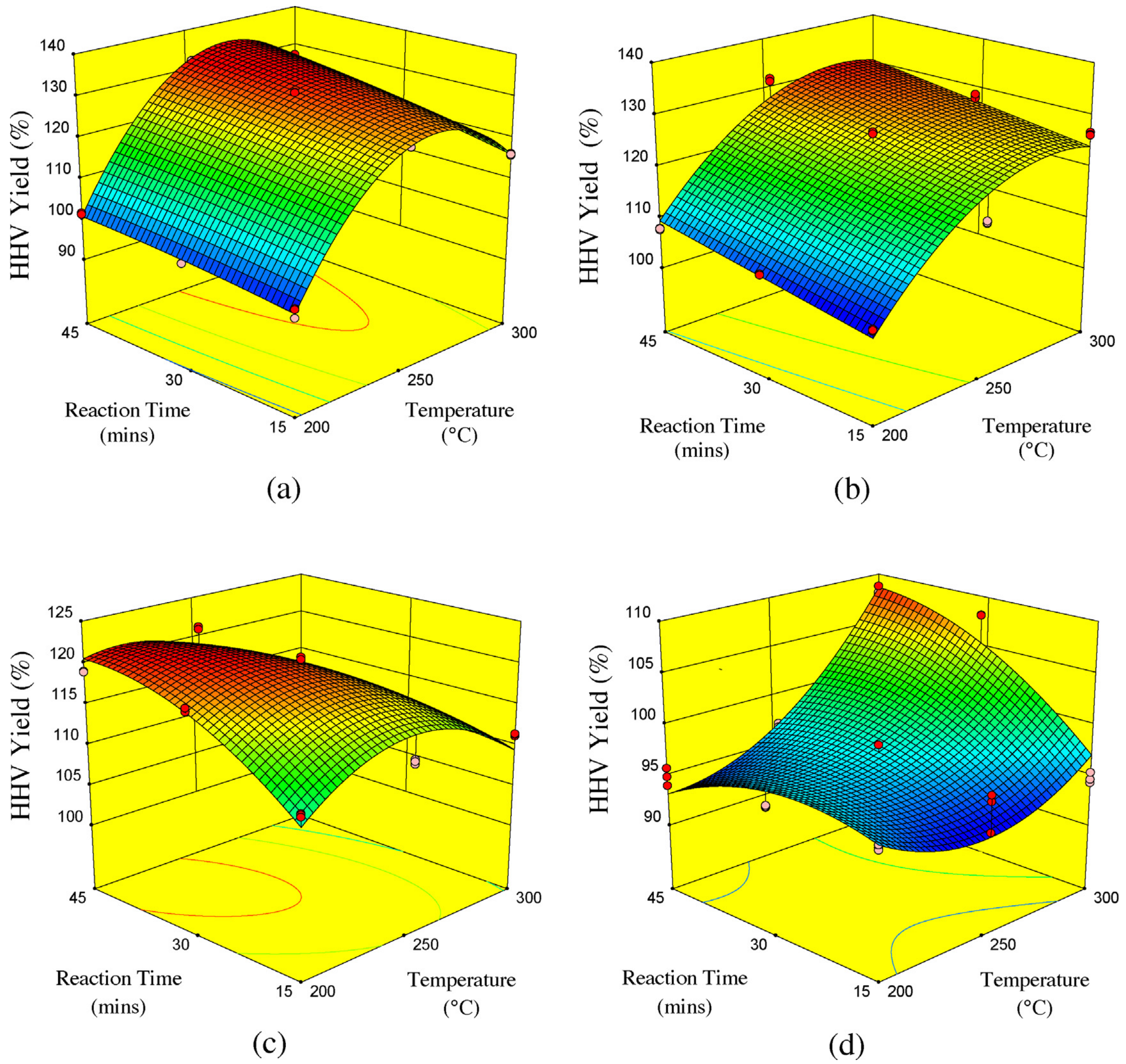

Fig. 1. Response surface and contour plot of torrefaction temperature vs. reaction time on the HHV yield for; (a) *Acacia* spp., (b) *Macaranga* spp., (c) EFB and (d) OPT.

combustible properties comparable to those of the fossil coal, leading to reduced costs and economic gains. Regarding fuel efficiency, it is necessary to increase the energy density (HHV yield) of the biomass, requiring a growth of the ratio between HHV and mass. The HHV yield per raw material indicates the total energy conserved in the torrefied biomass. The HHV yield was calculated from the weight loss and HHV using Eq. (1) and expressed as a percentage of the HHV of untreated lignocellulosic biomass: HHV_f denotes specific energy content of lignocellulosic biomass after torrefaction and HHV_i, specific energy content of biomass before torrefaction.

$$\text{HHV yield}(\%) = (100 - \text{weight loss}) \times \left(\frac{HHV_f}{HHV_i}\right) \quad (2)$$

The HHV yields of the lignocellulosic biomass are listed in Table 3. The results conclude that almost all the HHV of the lignocellulosic biomass is retained in the solid product. It is important to note that the energy yield can be greater than 100% as it is based on HHV and not enthalpy; energy is still conserved in the process. OPT displays the lowest energy yield, dropping to as low as 95% at 250 °C.

HHV yield was fitted to the response surface model provided by the mathematic models in Table 4, in order to analyze the effect of the torrefaction factor on HHV yield. Y_1 is the predicted HHV yield (%), where X_1 and X_2 denote temperature and reaction time. To fit the response function and experimental data, regression analysis was performed and the second order model for HHV yield was evaluated by analysis of variance (ANOVA) which is presented in Table 5. Table 5 shows the ANOVA of the quadratic model adjustment, where the total error was classified into lack of fit and pure error. The *F*-value estimated using the experimental data corresponded to the total residual and lack-of-fit values, respectively, and was lower than the tubular F value. This indicates that the models were significant in the region studied. The mathematic models (Table 4) shows in good correlation with the actual data as justified by the relatively high *R*-squared value. For all the models, the *p*-value was <0.0001, which shows that the models were strongly significant at the 99% confidence level. According to the models (Table 5), torrefaction parameters; temperature, time, temperature × time showed a significant value ($p \leq 0.05$) except *Macaranga* spp. No interaction was observed between the two parameters (temperature and time) used in torrefaction of *Macaranga* spp. on the HHV yield, indicated that different levels of each torrefaction parameter affect the HHV yield independently. Whereas, time squared ($Time^2$) were not a significant parameter ($p > 0.05$) for both models of *Acacia* spp. and *Macaranga* spp.

The HHV yield results obtained for all runs of the experimental design for the lignocellulosic biomass were represented by response surfaces (Fig. 1). The responses for HHV yield from the torrefactions were depicted as three dimensional surface plots of two factors; temperature and reaction time with their corresponding

Table 6
Predicted optimal torrefaction temperature and reaction time with the corresponding weight loss and HHV.

Lignocellulosic biomass	Temperature (°C)	Reaction time (min)	Weight loss (%)	HHV (MJ/kg)	HHV yield (%)
Acacia spp.	260	30	8.96	26.50	131.47
Macaranga spp.	280	45	13.84	27.30	131.04
EFB	230	40	9.91	24.30	121.15
OPT	300	45	14.00	21.82	109.23

contours plots. The strength of the effect of these two parameters on HHV yield is revealed more clearly by the surface plot. Temperature had a higher impact on the HHV yield of torrefied lignocellulosic biomass, while the effect of reaction time was considerable lower. Fig. 1 also illustrated the predicted peak in HHV yield at the optimum condition.

Numerical optimization using the response optimizer in Design-Expert Version 8.0.1 was carried out to find conditions for maximized the HHV yield. The approach allowed to predict the optimal torrefaction conditions for maximized high energy density. The predicted values of torrefaction temperature and reaction time obtained from mathematic models in Table 4 with the expected corresponding weight loss and HHV are shown in Table 6. In the range of temperature and reaction time studied here, EFB tends to carbonize easier during torrefaction than the other biomass types used in this study. Therefore, when the weight loss and HHV of biomass by torrefaction is considered, applying a mild torrefaction treatment (230 °C, 40 min) for EFB is appropriate for the maximized HHV yield in the biofuel. The optimum torrefaction conditions for high HHV yield from OPT is temperature of 300 °C and 45 min treatment. On the other hand, in order to obtain high HHV yield from *Acacia* spp. and *Macaranga* spp., the optimum torrefaction condition of 260 °C for 30 min and 280 °C for 45 min, respectively was predicted.

The validity of these results was verified and confirmed through performing torrefaction treatment for each of the lignocellulosic biomass types under the optimized conditions. For respective types of lignocellulosic biomass, the optimization experiment gave results of HHV and weight loss as follows: 25.96 MJ/kg, 9.12% for *Acacia* spp.; 27.19 MJ/kg, 13.41%, for *Macaranga* spp.; 23.00 MJ/kg, 9.55%, for EFB and 22.22 MJ/kg, 14.94% for OPT. These experimental findings were in close agreement with the model prediction.

4. Conclusion

Biomass can be upgraded and used as a fuel by torrefaction process. The torrefied biomass is more suitable than raw biomass in terms of HHV, physical and chemical properties. The optimum conditions for the torrefaction of selected fast growing species grown in Malaysia and oil palm biomass were investigated with respect to the reaction temperature and time. While the reaction temperature had a strong impact on the HHV yield of torrefied lignocellulosic biomass, the effect of reaction time was considerably lesser. As a whole, the torrefaction at 200 °C just degraded a small amount of holocellulose and gave a mediocre effect on improving the energy properties. When biomass underwent the torrefaction at temperatures above 250 °C, large amount of hemicelluloses and cellulose were degraded which contributed to the increment of acid-insoluble material. Weight loss and HHV vary for different biomass types at the same operating conditions as the polymeric composition and reactivity differ. In consequence, each biomass type has its own set of operating conditions to achieve a high density solid biofuel. The developed model for optimizing the torrefaction condition for maximum HHV yield (maximum HHV with minimum weight loss) was proved to be correct. The results of this study could be used as a guide for the production of high density solid biofuel from lignocellulosic biomass available in large amounts in tropical regions.

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